

DOCKET NO: 274437US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :  
KOZO TAKATSU, ET AL. : EXAMINER: SINGH, PREM C.  
SERIAL NO: 10/540,881 :  
FILED: JANUARY 18, 2006 : GROUP ART UNIT: 1797  
FOR: METHOD FOR REMOVING :  
SULFUR COMPOUND IN  
HYDROCARBON-CONTAINING GAS

APPEAL BRIEF

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal from the Examiner's Final Rejection dated September 1, 2010, of Claims 1-4 and 7-17. A Notice of Appeal was filed on February 1, 2011. This version of the Appeal Brief is in response to the Notification of Non-Compliant Appeal Brief dated May 9, 2011.

**I. REAL PARTY IN INTEREST**

The real party in interest is Idemitsu Kosan Co., Ltd., having an address of 1-1, Marunouchi 3-Chome, Chiyoda-Ku, Tokyo, Japan, by virtue of the assignment recorded July 30, 2007, at Reel/Frame 017474/0695.

## **II. RELATED APPEALS AND INTERFERENCES**

Appellants, Appellants' legal representative and their assignee are not aware of any other appeals or interferences which will directly affect or be directly affected by or having a bearing on the Board's decision in this appeal.

## **III. STATUS OF CLAIMS**

The appealed claims are Claims 1-4 and 7-16. Claims 1-4 and 7-16 stand twice rejected.

## **IV. STATUS OF AMENDMENTS**

No Amendment under 37 C.F.R. §1.116 was filed. A Request for Reconsideration was filed August 24, 2010.

## **V. SUMMARY OF CLAIMED SUBJECT MATTER**

It is preliminarily noted that the numbers in brackets refer to page and line number in the original specification as filed.

**Claim 1** provides a method for removing sulfur compounds [page 3, lines 11-16] contained in a hydrocarbon-containing gas [page 2, lines 8-18] comprising feeding a hydrocarbon-containing gas to a desulfurizing bed to remove sulfur compounds contained in the hydrocarbon-containing gas [page 6, lines 2-10], wherein the desulfurizing bed comprises: a desulfurizing agent A [page 10, lines 8-14] comprising at least one metal component selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, an alkali metal, an alkaline earth metal and a rare earth metal carried on a zeolite [page 10, line 19 to page 11, line 4]; and

a desulfurizing agent B [page 11, line 21, to page 12, line 12] comprising at least one selected from the group consisting of cerium oxide and a metal component-carried on a porous inorganic oxide,  
wherein

the metal component-carried on a porous inorganic oxide is one selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, Al, Si, an alkali metal, an alkaline earth metal and a rare earth metal [page 12, lines 5-13], and

the porous inorganic oxide is at least one selected from the group consisting of alumina, silica, silica-alumina and cerium oxide [page 12, lines 13-25].

Claims 3-4 and 7-16 depend from and stand or fall with Claim 1.

Claim 2 provides a method for removing sulfur compounds contained in a hydrocarbon-containing gas as described in claim 1, wherein

the desulfurizing agent A has a higher desulfurizing performance to sulfides and disulfides than that of the desulfurizing agent B, and

the desulfurizing agent B has a higher desulfurizing performance to carbonyl sulfide than that of the desulfurizing agent A [page 6, lines 12-20].

## **VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

Claims 1-4 and 7-16 stand twice rejected as being obvious under 35 U.S.C. §103 (a) over Takashi et al.(JP 2001-278602)(Takashi) in view of Satokawa et al. (U.S.2001/0014304)( Satokawa).

## **VII. ARGUMENT**

### **Rejection of Claims 1, 3-4 and 7-16 under 35 U.S.C. §103 (a) over Takashi in view of Satokawa**

The claimed invention provides a method for removing sulfur compounds, including carbonyl sulfide [page 1, lines 1-5] from fuels which may be consumed in a fuel cell. The claimed method comprises feeding a hydrocarbon-containing gas to a desulfurizing bed, containing 1) a desulfurizing agent A comprising at least one metal component selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, an alkali metal, an alkaline earth metal and a rare earth metal carried on a zeolite; and 2) a desulfurizing agent B comprising at least one selected from the group consisting of cerium oxide and a metal component-carried on a porous inorganic oxide. The metal component is one selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, Al, Si, an alkali metal, an alkaline earth metal and a rare earth metal, and the porous inorganic oxide is at least one selected from the group consisting of alumina, silica, silica-alumina and cerium oxide.

Takashi is directed to a method for removing sulfur contaminants from **petroleum hydrocarbon oils** (Abstract), by contacting the petroleum hydrocarbon with a first desulphurization agent which is a halide compound supported on a porous substrate [0005] and [0007] (17<sup>th</sup> group element, preferably Bromine). Additionally the oil may be treated with a second devulcanizing agent which may be of any kind. Examples of the second agent include hydrodesulfurization over a catalyst such as Co-Mo/alumina or Ni-Mo/alumina or adsorption by an agent being a metal on a porous support [0010].

Takashi describes the function of the halogen (Bromine) is to form a sulfur bromide which is more easily adsorbed than benzothiophenes and dibenzothiophenes and therefore removed by a porous adsorbent [0007].

Appellants note that Takashi describes U.S. 4,188,285, U.S. 5, 807,475 and 5,935,422, among others, [0003] as references describing adsorption methods based on a

zeolite which **do not adequately remove sulfur compounds from a fuel to allow utility for a fuel cell**. Appellants further note that each of the three cited references describes methods to remove sulfur compounds from a hydrocarbon stream with metal exchanged zeolite systems. To overcome the inadequacy of the described metal exchanged zeolite systems, Takashi describes the use of a Group 17 compound such as HBr. Therefore, Appellants submit that Takashi teaches the HBr system is superior to a metal exchanged zeolite system **for removal of benzothiophenes and dibenzothiophenes from petroleum hydrocarbon oils**.

Takahashi further describes that the HBr/porous inorganic system may be supplemented with a second devulcanizing agent and further indicates any type of desulfurization system may be used for the secondary treatment [0010]. Such system may be a hydrodehydrodesulfurization or an adsorbent system.

Satokawa describes a zeolite ion-exchanged with one or more transition metals as an adsorbent for removing sulfur compounds such as sulfides, thiophenes or mercaptans, from **fuel gas** (Abstract). Appellants submit that the secondary reference is directed to the removal of sulfur compounds from a gaseous mixture. Although, Satokawa describes that thiophenes may be a component of the fuel gas mixture, Appellants note that thiophene has a boiling point of 84.4°C, while the boiling points of benzothiophene and dibenzothiophene are 221 °C and 332-333 °C and therefore, benzothiophene and dibenzothiophene are not likely to be present in a fuel gas mixture such as city gas, natural gas or LP gas.

Appellants further submit that Satokawa is directed to removal of sulfides from a gaseous mixture, while Takashi is directed to solving a specific problem related to removal of benzothiophene and dibenzothiophene from petroleum hydrocarbons, materials highly unlikely to be present in the gaseous fuels to which the Satokawa method is directed. Appellants submit that Takashi describes this difference in [0002] and then further describes in [0003] that metal exchanged zeolites as described by Satokawa are not effective to remove benzothiophene and dibenzothiophene from a hydrocarbon petroleum. Appellants further

submit that as described, Takashi teaches away from metal exchanged zeolites as being effective for removal of benzothiophene and dibenzothiophene from a petroleum hydrocarbon.

In contradiction to the described differences in technology and the teaching away from the use of metal exchanged zeolites by the primary reference, the Office alleges that “a Group 17 element supported on a zeolite, used in the Takashi invention, and transition metal as supported on a zeolite, used in Satokawa invention, are functionally similar in the desulfurization process” (Official Action dated September 1, 2010, page 4, third paragraph). Appellants submit that as described above, the Office’s statement is erroneous. The two methods are not functionally similar and one of ordinary skill in the art would not look to the description of Satokawa to address removal of benzothiophene and dibenzothiophene from a petroleum hydrocarbon. Takashi indicates that such treatment is not effective and for that reason employs the HBr method.

Appellants submit that the Office continues to fail to recognize the mode of operation of the Takashi agent which is described in [0007] as follows (machine translation):

This bromine reacts with sulfur and changes to a sulfur bromide with the adsorption capacity force higher than benzothiophene and dibenzothiophene.

In contrast, the agent described by Satokawa is a simple adsorbent which does not provide for formation of a sulfur bromide and therefore, replacement of the Takashi Group 17 carrying zeolite with the agent of Satokawa would change the principle of operation of the primary reference and as described by Takashi is ineffective for the removal of benzothiophene and dibenzothiophene from a petroleum hydrocarbon.

Appellants respectfully submit that the Office has not provided any reasonable explanation with a technical underpinning of why one of ordinary skill in the art, at the time of the present invention would have disregarded the teaching of the primary reference and

substituted the adsorbent of Satokawa for the HBr system of Takashi. Moreover, the Office has not explained how or why one of ordinary skill would have combined the Satokawa system with a metal on a porous support according to the present invention. The Office appears not to recognize that Takashi describes both hydrodesulfurization and adsorption methods (see Satokawa, [0007] for an explanation of the methods) and does not explain how or why one of ordinary skill would have selected the particular combination according to the present invention.

For all the above reasons. Appellants respectfully submit that the rejection of Claims 1-4 and 7-16 under 35 U.S.C. §103 (a) over Takashi in view of Satokawa is improper and should be reversed.

Rejection of Claim 2 under 35 U.S.C. §103 (a) over Takashi in view of Satokawa

Claim 2 depends from Claim 1 and includes all the description of the independent claim. Claim 2 further describes that the desulfurizing agent A has a higher desulfurizing performance to sulfides and disulfides than that of the desulfurizing agent B, and the desulfurizing agent B has a higher desulfurizing performance to carbonyl sulfide than that of the desulfurizing agent A. Appellants submit that both Takashi and Satokawa are silent with respect to removal of carbonyl sulfide. Applicants further note that Satokawa corresponds to JP 2001-286753 which is described in the specification on page 4, lines 1-8, as not effective for the absorption of carbonyl sulfide.

Appellants submit that the Office has basically alleged that since the combination of the references describes the same agents as the invention, the relationship according to Claim 2 would be satisfied. However, as Appellants have shown the two references cannot be properly combined to obtain the claimed invention. Moreover, Applicants submit that the

data shown in the following Table demonstrates a **synergistic improvement** in desulfurization is obtained by the combination of agents A and B according to the invention. The data for Example 1 and Comparative Examples 1 and 2 are shown below:

Example	COS	DMS	TBM	DMDS
1	6	11	>15	15
Comp. 1	0	11	>15	9
Comp. 2	1	0	>15	3

In the table, COS is carbonyl sulfide, DMS is dimethyl sulfide, TBM is t-butylmercaptan and DMDS is dimethyl disulfide. The numbers in the table show the hours of catalyst effectiveness in removing the listed contaminant from a feed stream containing the contaminants. Appellants note that Comp. 1 corresponds to the desulfurizing agent described by Satokawa and Example 1 is a catalyst according to the invention which is a combination of the two comparative example agents.

As indicated in the table, the desulfurizing agent according to the invention is effective for removal of COS and DMDS for significantly longer time than would be expected based on the individual components and shows a synergistic improvement. Appellants submit that such a showing of greater than expected results is evidence of nonobviousness which supports the patentability of the present invention.

In response to such showing, the Office has alleged that Takashi recognizes such a synergistic effect (Official Action dated September 1, 2010, page 10, lines 3-9). Appellants respectfully submit that such a position is irrational and without technical support. As described above, Takashi teaches that metal exchanged zeolite is not effective for the



problem addressed by that reference and therefore, teaches away from such an agent as a primary adsorbent. Moreover, Takashi does not disclose or suggest the combination of two adsorbent systems according to the present invention and Takashi is silent with respect to carbonyl sulfide. Appellants submit that the reference cannot recognize a synergistic effect for a combination of adsorbents it does not describe for removal of a contaminant it does not recognize.

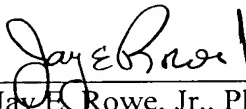
Accordingly, and in view of all the above, Appellants submit that the rejection of Claim 2 under 35 U.S.C. §103 (a) over Takashi in view of Satokawa is improper and should be reversed.

#### CONCLUSION

For all the above reasons, Appellants submit that all rejections of record should be reversed.

Respectfully submitted,

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### VIII. CLAIMS APPENDIX

Claim 1 (Rejected): A method for removing sulfur compounds contained in a hydrocarbon-containing gas comprising feeding a hydrocarbon-containing gas to a desulfurizing bed to remove sulfur compounds contained in the hydrocarbon-containing gas, wherein the desulfurizing bed comprises:

a desulfurizing agent A comprising at least one metal component selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, an alkali metal, an alkaline earth metal and a rare earth metal carried on a zeolite; and

a desulfurizing agent B comprising at least one selected from the group consisting of cerium oxide and a metal component-carried on a porous inorganic oxide, wherein

the metal component-carried on a porous inorganic oxide is one selected from the group consisting of Ag, Cu, Ni, Zn, Mn, Fe, Co, Al, Si, an alkali metal, an alkaline earth metal and a rare earth metal, and

the porous inorganic oxide is at least one selected from the group consisting of alumina, silica, silica-alumina and cerium oxide.

Claim 2 (Rejected). The method for removing sulfur compounds contained in a hydrocarbon-containing gas as described in claim 1, wherein

the desulfurizing agent A has a higher desulfurizing performance to sulfides and disulfides than that of the desulfurizing agent B, and

the desulfurizing agent B has a higher desulfurizing performance to carbonyl sulfide than that of the desulfurizing agent A.

Claim 3 (Rejected): The method for removing sulfur compounds contained in a hydrocarbon-containing gas as described in claim 1, wherein a volume ratio of the desulfurizing agent A to the desulfurizing agent B in the desulfurizing bed is 0.1 : 0.9 to 0.9 : 0.1.

Claim 4 (Rejected): The method for removing sulfur compounds contained in a hydrocarbon-containing gas as described claim 1, wherein zeolite in the desulfurizing agent A has a beta (BEA) and/or faujasite (FAU) structure.

Claims 5 and 6 (Canceled).

Claim 7 (Rejected): The method for removing sulfur compounds contained in a hydrocarbon-containing gas as described claim 1, wherein a temperature of the desulfurizing bed is -20 to 100°C.

Claim 8 (Rejected): A hydrocarbon-containing gas for a fuel cell obtained according to the method of claim 1, wherein a concentration of carbonyl sulfide is 0.1 weight ppm or less.

Claim 9 (Rejected): The hydrocarbon-containing gas for a fuel cell as described in claim 8, wherein the hydrocarbon-containing gas for a fuel cell is at least one selected from natural gas, city gas, LPG, a naphtha fraction and dimethyl ether.

Claim 10 (Rejected): A method for removing sulfur compounds contained in a hydrocarbon-containing gas, wherein a desulfurizing agent comprising at least zeolite is used to remove sulfur compounds contained in the raw material hydrocarbon-containing gas for a fuel cell as described in claim 8.

Claim 11 (Rejected): A method to produce hydrogen for a fuel cell, comprising:  
removing sulfur compounds contained in a hydrocarbon-containing gas by the method  
as described in claim 1, and

then contacting the hydrocarbon-containing gas from which the sulfur compounds  
have been removed with one selected from the group consisting of a partial oxidation  
reforming catalyst, an autothermal reforming catalyst and a steam reforming catalyst.

Claim 12 (Rejected): The method for producing hydrogen for a fuel cell as described  
in claim 11, wherein the partial oxidation reforming catalyst, the autothermal reforming  
catalyst or the steam reforming catalyst is a ruthenium base or nickel base catalyst.

Claim 13 (Rejected): A method for producing hydrogen for a fuel cell, wherein the  
hydrocarbon-containing gas as described in claim 8 is a raw material.

Claim 14 (Rejected): A method for producing hydrogen for a fuel cell, comprising:  
removing the sulfur compounds contained in the hydrocarbon-containing gas as  
described in claim 8, and then

contacting the hydrocarbon-containing gas from which the sulfur compounds have  
been removed with one selected from the group consisting of a partial oxidation reforming  
catalyst, an autothermal reforming catalyst and a steam reforming catalyst.

Claim 15 (Rejected): The method for producing hydrogen for a fuel cell as described  
in claim 14, wherein the partial oxidation reforming catalyst, the autothermal reforming  
catalyst or the steam reforming catalyst is a ruthenium base or nickel base catalyst.

Claim 16 (Rejected): The method for removing sulfur compounds contained in a hydrocarbon-containing gas as described in claim 1, wherein

the desulfurizing agent A comprises zeolite having a beta (BEA) and/or faujasite (FAU) structure and at least one metal component selected from the group consisting of Ag and Cu, and

the desulfurizing agent B comprises at least one selected from the group consisting of cerium oxide and a metal component-carried on a porous inorganic oxide, wherein

the metal component is at least one selected from the group consisting of Ag, Cu and Ni, and

the porous inorganic oxide is at least one selected from the group consisting of alumina, silica-alumina and cerium oxide.

IX. EVIDENCE APPENDIX

None

X. RELATED PROCEEDINGS APPENDIX

None